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Measurements of Gross α- and β-Activities of Archived $PM_{2.5}$ and PM_{10} Teflon Filter Samples

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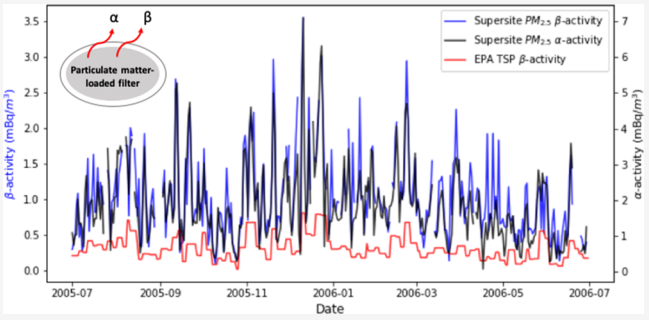
ABSTRACT: The adverse effects of ambient particulate matter (PM) on human health have been well demonstrated, but the underlying properties responsible for its toxicity are still unclear. We hypothesized that particulate radioactivity, which is due to the attachment of radioactive nuclides on particle surfaces, may be responsible for part of PM toxicity. We measured the gross α- and β-activities for daily $PM_{2.5}$ and PM_{10} filters collected at the Harvard Supersite in downtown Boston from 2005 to 2006 and calculated the radioactivities at the time of air sampling retrospectively based on a previously established formula. We examined the relationship between different radioactivities and compared our measurements to those measured at the Boston EPA RadNet Station. The results showed that the majority of PM_{10} radioactivity is associated with that of $PM_{2.5}$ samples for both α-activity (98%) and β-activity (83%). A strong linear relationship was observed between the α- and β-activities for both $PM_{2.5}$ [slope = 0.47 (±0.03); p-value < 0.0001] and PM_{10} [slope = 0.46 (±0.09); p-value < 0.0001] samples. Measurements at the Harvard Supersite and at EPA RadNet sites are highly correlated for both α-activities [slope = 0.17 (±0.02), p-value < 0.0001] and β-activities [slope = 0.30 (±0.05), p-value < 0.0001]. Additionally, we identified several significant predictors for $PM_{2.5}$ α-activities. This novel method we developed to measure α- and β-activities from archived filters will make it possible to assess the retrospective particle radioactivity exposure for future epidemiological studies.



INTRODUCTION

Ambient air pollution accounts for approximately 4.2 million deaths per year due to stroke, heart disease, lung cancer, and chronic respiratory diseases. Among all the air pollutants, particulate matter (PM) contributes the strongest evidence for public health concern.[1] In addition, ambient PM remains the major source of indoor PM, contributing from 23 to 67% to the indoor particle concentrations.[2−6] While the adverse effects of ambient PM on multiple health outcomes have been well-demonstrated,[7−11] the underlying PM properties responsible for its toxicity are still under intense scientific investigation. PM can serve as a surface platform to mediate and catalyze chemical processes that are important for human health.[12,13] To date, studies have focused on the potential role of particle mass, number, solubility, morphology (structure, porosity, size distribution, surface area, etc.), and chemical composition (organic carbon/elemental carbon, sulfate, metals, etc.).[14−17] Recently, we hypothesized that particulate radioactivity (PR), which is due to the attachment of radioactive nuclides on particle surfaces, may be responsible for part of PM toxicity. In this case, PM is acting as a vector for radioactive isotopes, which carries them into the human body when inhaled.[18]

In the absence of anthropogenic sources of radioactivity, radon (^{222}Rn) and its decay products serve as the primary source of ionizing radiation exposure. ^{222}Rn is a naturally occurring radioisotope from the decay of ^{238}U and ^{232}Th, with a half-life of 3.8 days. The decay of ^{222}Rn produces progeny in the form of charged radioactive ions, which are subsequently neutralized or aggregated with water molecules or other gaseous species.[19] Eventually, the majority of these radioactive ions attach onto airborne particles and get into the human body by inhalation. Most of the inhaled ^{222}Rn gas is quickly exhaled, but the radioactive particles can deposit onto bronchial epithelial cells and further travel to other tissues and organs through the circulatory system. This can lead to irradiation in the biological systems as these radioisotopes emit high-energy α- and β-particles, inducing serious adverse health effects.[8,20−23] As PM and ^{222}Rn are ubiquitous, it is important

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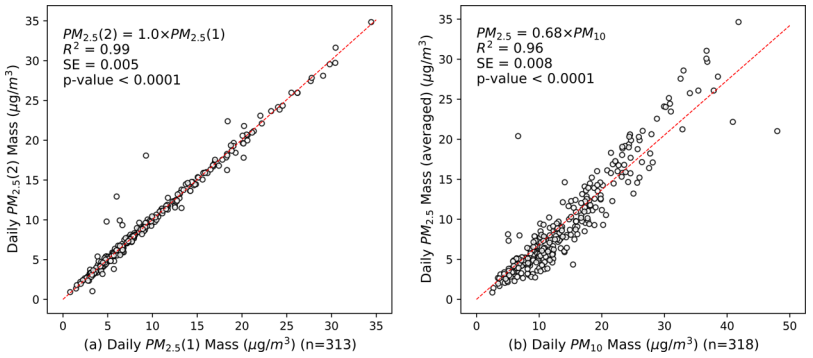

Figure 1. Comparisons between mass concentrations (a) of the duplicate $PM_{2.5}$ (1 and 2) samples and (b) PM_{10} and $PM_{2.5}$ samples (averaged).

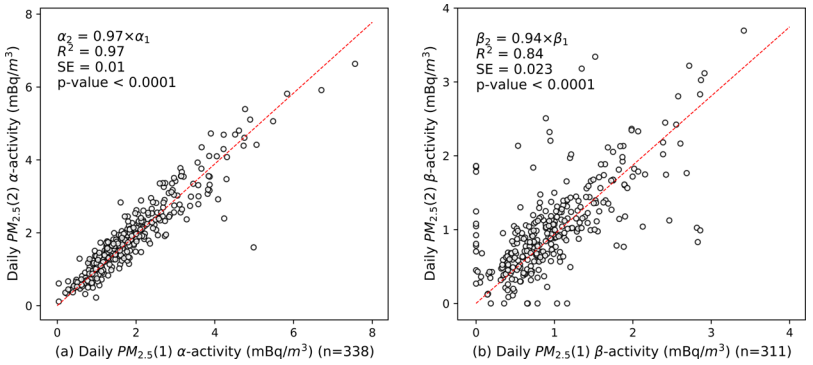

Figure 2. Comparisons between the two duplicate $PM_{2.5}$ (1 and 2) samples for (a) α-activities and (b) β-activities.

to measure the radioactivity of PM to estimate radiation exposures.

PM gross α- and β-activities are easy to measure using nondestructive methods.[24] Systematic measurement of gross α- and β-activities can be used to investigate trends of environmental radioactivity levels. Some studies have assessed the variations of gross α- and β-atmospheric activities and have identified the most important variables influencing them, for example, meteorological parameters.[25−27] Several other studies have identified and quantified specific radionuclides, such as ^{7}Be, ^{210}Pb, ^{40}K, and so forth, which are responsible for particle radioactivity.[28−30] However, few studies have examined relationships between ambient particle radioactivity and other particle physicochemical characteristics such as the mass concentration, size, and chemical composition.[31]

In this study, we measured gross α- and β-activities from archived $PM_{2.5}$ and PM_{10} Teflon filter samples collected over a period of 1 year in Boston, MA. We report the activity levels and characterize their seasonal patterns, $PM_{2.5}/PM_{10}$ ratios, and relationships with some meteorological parameters such as temperature, precipitation, wind speed, and relative humidity. Using this novel method, we are able to assess population exposure to particulate radioactivities retrospectively for up to 20 years, which provides us with a new approach for exposure assessment and a new angle for future epidemiological studies looking into the adverse health impact of particulate radioactivities.

METHODS

$PM_{2.5}$ and PM_{10} Sampling. Ambient $PM_{2.5}$ and PM_{10} samples were collected at the Harvard air-monitoring Supersite during a 1 year period from July 1, 2005 to June 30, 2006. The Supersite is located on the roof of the Countway Library at the Harvard Medical School in downtown Boston, MA. PM samples were collected for 24 h using two Harvard impactor samplers with cut points of 2.5 and 10 μm, respectively, operating at a flow rate of 10 L/min. PM samples were collected on 37 mm Teflon filters. Every day, two duplicate $PM_{2.5}$ samples and one PM_{10} sample were collected. The filters were pre- and postweighted using an electronic microbalance (Mettler MT-5, Columbus, OH) in a temperature- and relative humidity-controlled environment to determine the PM mass concentration.[32,33]

Particulate Radioactivity. After storage for approximately 13 years, we measured particle α- and β-activities from long-lived ^{222}Rn progeny (^{210}Pb to ^{210}Bi to ^{210}Po) for the PM filters using a low background gas proportional counter (model LB4200, Canberra Industries, Inc., Meriden, CT) with a P10 carrier gas (10% methane balanced with argon) installed at the Harvard T.H. Chan School of Public Health. Based on preliminary tests for counting times between 300 and 1200 min, we selected an optimum time of 600 min to accurately measure the relatively low concentrations of α- and β-activities. The counter was calibrated every 2 weeks with a 0.0518 μCi NIST-traceable ^{210}Po α-source and a 0.00516 μCi NIST-traceable ^{90}Sr β-source on 5.7 cm planchets. The counting efficiency of α- and β-activities determined was 40 ± 1% (mean ± standard deviation) and 50.3 ± 0.8%, respectively. The background level was below 0.1 cpm (count per minute). The detection limit (LOD) is 0.219 mBq/m^3 for α-activity and 0.355 mBq/m^3 for β-activity, with a counting time of 600 min and a sample volume of 14.4 m^3.[34] We applied a background correction procedure to each of the gross α- and β-activity measurements before conducting statistical analysis. Most of the α- and β-measured activities were substantially higher than the detection limit, as reported in Table 1.

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Several different Rn progenies attached to PM were collected on filters during sampling. As these collected radionuclides decay into radioactive products, the products contribute their own activities to those already on the filter. The total activity on a filter at any time after collection is a function of the types of radionuclides, their air concentrations, and the sampling rate and duration. The α-activity initially decreases as the shorter-lived Rn progeny decay to β-emitting ^{210}Pb, whose half-life is about 22 years. After reaching a minimum at about 6 days, α-activity then increases for approximately 3 years as ^{210}Pb decays to a short-lived β-emitter, ^{210}Bi, which then decays to ^{210}Po. ^{210}Po emits α-particles and finally decays to stable ^{206}Pb.[35] At around 3 years, a transient equilibrium between ^{210}Pb and ^{210}Po is reached, after which the α-activity declines exponentially controlled by the ^{210}Pb decay rate.[36,37] Furthermore, calculations have shown that after about 1 year, essentially all the α-activity remaining on the filter is from ^{210}Po, whose source was ^{210}Pb present in the air at the time of sampling.[38] Therefore, we could use the α-activity of the filter measured at counting to calculate the long-lived α-activity on the filter at the time of sampling. This can be accomplished using the following relationship, which is based on Sheets and Thompson's original equation estimating the atmospheric concentration of ^{210}Pb[38]

$$C = \frac{A_{\alpha(t)} \cdot e^{\lambda t}}{Q}$$

where $A_{\alpha(t)}$ is the α-activity on the filter at counting (mBq), λ is the decay constant for ^{210}Pb (8.51×10^{-5} d^{-1}), t is the duration in days between PM sampling and α-activity measurement, and Q is the air sample volume (m^3). Because of the complex mixture of β-sources,[39] there is no simple formula to calculate the total β-activity level at the time of sample collection. Consequently, the best estimate for β-activity was determined using the same equation, assuming that the decay of ^{210}Pb, which is the species with the longest half-life, dominated the PR at the time of sample collection.

EPA Gross β-Activity Data. In the United States, the Environmental Protection Agency's (EPA's) Radiation Network (RadNet) monitoring program provides measurements of environmental radiation. The purpose of RadNet is to detect higher than normal radiation levels when a radiological incident occurs, as well as to track normal background radiation levels.[40] At each air monitoring site, a high-volume air sampler is used to collect total suspended particles (TSP) on a 4 in. diameter polyester fiber filter. Samples are collected approximately twice a week and are sent to the National Analytical Radiation Environmental Laboratory (NAREL) for the measurement of gross β-activity after 5−10 days following the collection date, thus allowing the shortest-lived ^{222}Rn progenies on the filters to fully decay.[41,42] We obtained gross β-activity data measured at the RadNet station located at Boston, collected from July 1, 2005 to June 30, 2006. As the sampling period for each sample is 3−5 days, we assigned the single measurement of β-activity to each day of the multiday sample.

Predictors of Particle Radioactivity. We were interested in the potential predictors of particle radioactivity. As meteorology plays an important role in the emission, transport, and fate of radionuclides,[25,27] we examined the effect of several meteorological parameters. Temperature, relative humidity, precipitation, and wind speed were obtained from the Boston Logan International Airport Weather Station.[43] The planetary boundary layer (PBL) height was obtained for the Supersite from the Physical Science Laboratory of National Oceanic Atmospheric Administration (NOAA-PSL).[44] We also included $PM_{2.5}$ and PM_{10} mass concentrations in the model. Additionally, particle radioactivity is influenced by the origin of air masses because of the higher emanation rate of ^{222}Rn from continental regions compared to the ocean.[45] For this reason, we calculated four daily 72 h back-trajectories (arrival time 06:00, 12:00, 18:00, and 24:00) at the Boston RadNet monitoring site using the hybrid single particle Lagrangian integrated trajectory (HYSPLIT) model.[46] The temporal proportion of these trajectories over the continent was used as a proxy for background sources. "0" indicates the maritime air mass, while "1" indicates the entirely continental air mass.

Statistical Analysis. We constructed linear regression models to characterize the seasonal patterns of PM gross α- and β-activities and their $PM_{2.5}/PM_{10}$ ratios, as well as their relationships between activities and potential predictors. As α- and β-activities are log-normally distributed, we applied logarithmic transformations before performing multivariate regression using Python 3.6[47] and RStudio 3.5.3.[48]

RESULTS

Measurement and Summary Statistics. The distributions of daily gross α- and β-activities are shown in Table 1. We conducted a descriptive analysis for the measurements of gross α- and β-activity levels of duplicate PM samples collected at the Boston Supersite, as well as for the gross β-activities from RadNet. The concentrations are expressed in mBq/m^3. The geometric means of α-activities of 686 duplicate $PM_{2.5}$ samples are 1.54 and 1.59 mBq/m^3, which are very similar to that estimated for the 390 PM_{10} samples, 1.58 mBq/m^3. The geometric means of β-activities of the 686 duplicate $PM_{2.5}$ samples are 0.37 and 0.54 mBq/m^3, which are slightly lower than the average β-activities of the 390 PM_{10} samples, 0.58 mBq/m^3. The average gross β-activities reported by RadNet TSP samples and for the same period is 0.27 mBq/m^3, which is smaller than our β-activity measurements. In total, we have 1007 samples above LOD for α-activity (89%) but only 806 samples above LOD for β-activity (71%). The gross α- and β-activity levels are both higher than those reported by previous studies.[25,26,29,49]

As shown in Figure 1, the particle mass concentrations of the two duplicated $PM_{2.5}$ samples are highly correlated, with a regression coefficient (R^2) of 0.99. In addition, the masses of $PM_{2.5}$ (1) and PM_{10} samples are in good agreement (R^2 = 0.96).

The relationships between activities are also good. The daily α- and β-activities of the $PM_{2.5}$ (1 and 2) samples are highly correlated with both regression coefficients and R^2 values approaching 1 (Figure 2a,b). These results demonstrate the high precision of the analytical method. For the subsequent data analysis, we averaged the measurements of two duplicated $PM_{2.5}$ (1 and 2) samples because of their high comparability. We also observed that β-activities are noisier compared to α-activities.

We regressed $PM_{2.5}$ α- and β-activities on the respective PM_{10} activities on a monthly basis to reduce noise from daily measurements. As shown in Figure 3a,b, $PM_{2.5}$ α-activity contributes over 98% of the PM_{10} α-activity (R^2 = 0.99). In addition, $PM_{2.5}$ β-activities contribute over 83% of the PM_{10} β-

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Table 1. Summary Statistics of Gross α- and β-Activity Levels during the Period of July 1, 2005 to June 30, 2006[a]

	$PM_{2.5}(1)$ α[b]	$PM_{2.5}(1)$ β	$PM_{2.5}(2)$ α[c]	$PM_{2.5}(2)$ β	PM_{10} α[d]	PM_{10} β	EPA β[e]
geom. mean[f]	1.54	0.37	1.59	0.54	1.58	0.58	0.27
SD	1.14	0.63	1.08	0.62	1.10	0.97	0.17
minimum	0.04	<LOD[g]	0.11	<LOD	0.04	<LOD	0.02
maximum	7.56	3.42	6.64	3.69	6.89	6.19	0.81

[a]All the activities are expressed in mBq/m^3. [b]α-activity of the first $PM_{2.5}$ sample between the two duplicates. [c]α-activity of the second $PM_{2.5}$ sample between the two duplicates. [d]α-activity of PM_{10} samples. [e]β-activity from EPA RadNet system. [f]Geom. mean = geometric mean. [g]<LOD denotes the measurements are below the limit of detection.

activities (R^2 = 0.97). These findings indicate that the majority of PM_{10} radioactivity is associated with that of $PM_{2.5}$.

A strong linear correlation was found between the daily gross α- and β-activities for both $PM_{2.5}$ (slope = 0.47, SE = 0.03, p-value < 0.0001) and PM_{10} (slope = 0.46, SE = 0.09, p-value < 0.0001) samples (Figure 4a,b). The ratio of the geometric means for the α- and β-activities was approximately 3.

Intercomparison between Activities Measured at the Supersite and RadNet Site. Figure 5 presents the times series of daily $PM_{2.5}$ α- and β-activities from the Harvard Boston Supersite and TSP β-activities from the EPA Boston RadNet station. Consistent with the above results, the average level of α-activities is about twice the level of β-activities, and both are much higher than the β-activities measured by RadNet. Figure 6 compares monthly TSP β-activities from the EPA Boston RadNet site to the respective $PM_{2.5}$ α- and β-activities from the Boston Harvard Supersite. As the EPA samples were collected during varying sampling periods (4−7 days), our comparisons are based on monthly means. The statistical analysis suggests a strong association for both α-activities (slope = 0.17, SE = 0.023, p-value < 0.0001) and β-activities (slope = 0.30, SE = 0.048, p-value ≤ 0.0001) (Figure 6).

Seasonal Variations of Particle Radioactivities. Figure 7 presents the seasonal patterns of particle mass, α-activities, and β-activities of $PM_{2.5}$. $PM_{2.5}$ mass is the lowest in spring and highest in summer; for radioactivity, spring has both the lowest average α-activities (1.47 mBq/m^3) and the lowest average β-activities (0.89 mBq/m^3). Winter has the highest average activities for both α (2.40 mBq/m^3) and β (1.35 mBq/m^3).

Predictors of Particle Radioactivity. We examined the effect of potential predictors using multivariate linear models. The following covariates were included in the analysis: temperature, barometric pressure, precipitation, wind speed, PBL height, $PM_{2.5}$ and PM_{10} mass, and temporal proportion of back-trajectories of air mass. α- and β-activities were log-transformed before modeling due to their skewness. For α-activity, all the regression coefficients are statistically significant at a 0.05 level (Table 2). For β-activity, the coefficients for pressure, precipitation, and PBL height were not significant. This analysis showed that $PM_{2.5}$ mass, atmospheric pressure, wind speed, and temporal proportion of back-trajectory over the continent are positively associated with radioactivity, while temperature, precipitation, and PBL height are negatively associated with radioactivity. For PM_{10}, the results are similar to those of $PM_{2.5}$ because of a high correlation between two measurements (see Supporting Information) and because most of the particle radioactivity is associated with $PM_{2.5}$, as shown above.

DISCUSSION

In this study, we used a novel method to measure the radioactivity of archived particle filters, which makes possible to assess past exposures for health-effect studies. Most of the daily $PM_{2.5}$ and PM_{10} samples measured were above the relatively low detection limits. The high correlations for the paired $PM_{2.5}$ samples indicate good precision of this method. In total, we measured α- and β-radioactivity for 686 $PM_{2.5}$ and 390 PM_{10} daily Teflon filter samples collected during the period from July 1, 2005 to June 30, 2006. We observed a high correlation between the measured α- and β-activities. In addition, our β-activity measurements were highly correlated with those previously reported for the RadNet system, although our measurements were higher.

Our findings suggest that most of the PM_{10} α- and β-activity measurements are associated with those of $PM_{2.5}$, over 98 and 83%, respectively. This is as expected, as previous studies have shown that the greatest activity fraction of ambient radioactive aerosols is in the accumulation mode[50,51] which, according to the general definition, comprises particle sizes between 0.1 and 1 μm.[52,53] In agreement with our measurements, a previous study on the activity size distributions of different natural radionuclides reported that most (93−96%) of the long-lived ^{222}Rn progeny (^{210}Pb and ^{210}Po) were attached to particles in the accumulation mode.[54] Considering that fine particles can penetrate deeply into the lungs and even enter the circulatory system,[55] they can be quite toxic because of their ability to carry most of airborne radionuclides into the body.

In our analyses of the association between daily α- and β-activities, we found a strong linear relationship for both $PM_{2.5}$ and PM_{10} sample types. The associations became stronger when we compared monthly averages instead of daily measurements, mainly due to the decrease in the measurement noise. In addition, we found an α-/β-activity ratio of approximately 3. In the absence of anthropogenic sources, the α-activity is mostly associated with the ^{222}Rn progeny, but β-activity has many other sources in the environment. Specifically, ^{210}Pb, which is a long-lived ^{222}Rn progeny, contributes a major part to particle β-activity. Other radionuclides, such as ^{40}K and ^{232}Th also contribute to particle β-activity.[28,56] Because the particle β-activity is associated with different sources, there is no single formula to calculate its level at the time when the sample was collected. For this study, we assumed that most of the β-activity is associated with ^{210}Pb because of its longest half-life. Despite this approximation, we found a strong relationship between our β-activities and those reported by RadNet. This provides us with confidence in using β-activities from the RadNet for our epidemiological studies.[18,57]

Both α- and β-activities of $PM_{2.5}$ samples were the lowest in spring, and highest in winter. This pattern was similar to that observed for PM_{10} samples (see Supporting Information).

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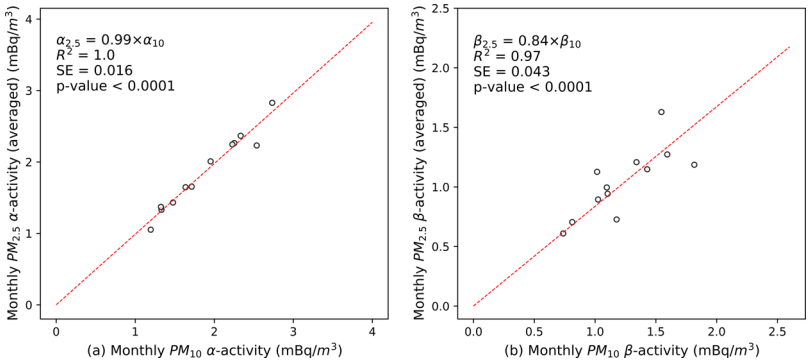

Figure 3. Comparisons between (a) monthly PM_{10} and $PM_{2.5}$ α-activities (averaged) and (b) monthly PM_{10} and $PM_{2.5}$ β-activities (averaged).

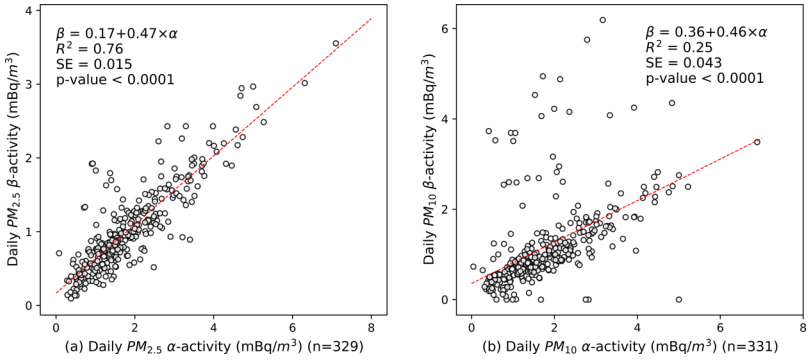

Figure 4. Comparisons between (a) daily $PM_{2.5}$ α- and β-activities (averaged) and (b) daily PM_{10} α- and β-activities (averaged).

Inconsistent seasonal patterns of particle α- and β-activities have been reported previously. Three studies focusing on the β-activity observed maxima and minima values during the summer and spring.[28,56,58] One study conducted in Salamanca, Spain, and another study conducted in Malaga, Spain, observed higher levels of α- and β-activities in summer and lower levels in winter, exactly opposite to our findings.[27,49] Other two studies conducted in Malaga, Spain, observed the highest value of α- and β-activities in summer, but lower values in January, April, and October, which indicate a more complicated seasonal variation.[25,26]

To examine the potential factors influencing PM radioactivity levels, we constructed multivariate regression models for $PM_{2.5}$ α- and β-activities. The factors we considered are the $PM_{2.5}$ mass; several meteorological parameters including the temperature, barometric pressure, precipitation, wind speed, and PBL height; and 72 h back-trajectories of air mass. The meteorological variables were selected based on previous studies in this field.[25,26,28−30,49,56] For the α-activity, we found significant associations for all the covariates; but for β-activity, pressure, precipitation, and PBL height were not significant, presumably due to a decrease in power considering the higher noise in β-activity measures as described above. $PM_{2.5}$ mass was positively associated with α- and β-activities, but there are no similar studies for us to compare with. Temperature was negatively associated with both α- and β-activities; corresponding somewhat to the seasonal pattern we found where activities peaked in winter. Pressure and wind speed had a positive impact, while the temperature, precipitation, and PBL height had a negative impact on radioactivity. To some extent, this corresponds to the seasonal pattern we observed. We used 72 h back-trajectories of air mass to estimate the proportion of air mass that originates from the continent rather than the ocean

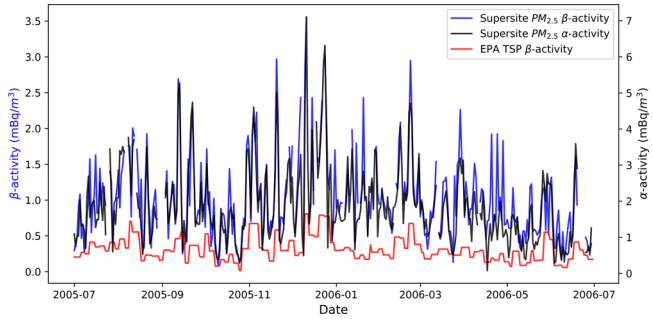

Figure 5. Time series of $PM_{2.5}$ α- and β-activities from the Boston Harvard Supersite and TSP β-activities from the Boston RadNet site.

before they arrive at Boston. We found this proportion to be positively associated with both α- and β-activities because air mass originating from the continent contains more ^{222}Rn and ^{222}Rn decay products as compared to those from the ocean.

As mentioned above, several previous studies have found that α- and β-activities were higher during summer, indicating a positive association with temperature that is consistent with our findings.[25−27,49,59] We found that both α- and β-activities were negatively associated with precipitation, as expected, because precipitation washes out particles from air.[60] Similarly, the PBL height was negatively associated with radioactivity, which is partially because of the decrease of PM concentration due to vertical mixing. In addition, ground-level Rn concentrations are known to decrease as the PBL height increases.[61] In fact, some atmospheric studies have used Rn concentrations as a surrogate of PBL height.[62,63] Pressure and wind speed were positively associated with α- and β-activity levels, but previous studies have shown both positive and negative association, making it difficult to draw a definite

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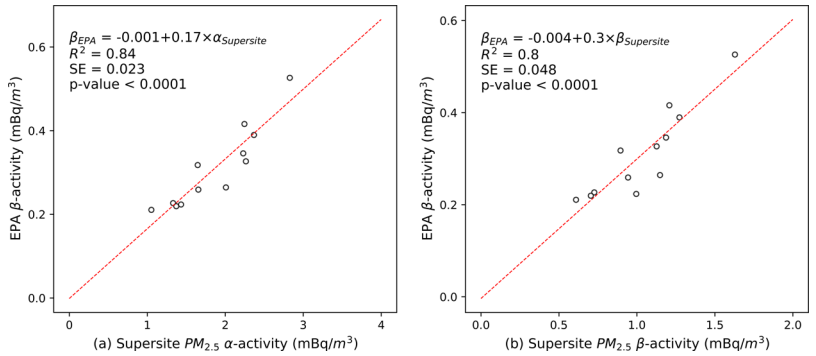

Figure 6. Comparisons between TSP β-activities from the Boston RadNet site and (a) $PM_{2.5}$ α-activities from the Boston Harvard Supersite and (b) $PM_{2.5}$ β-activities from the Boston Harvard Supersite.

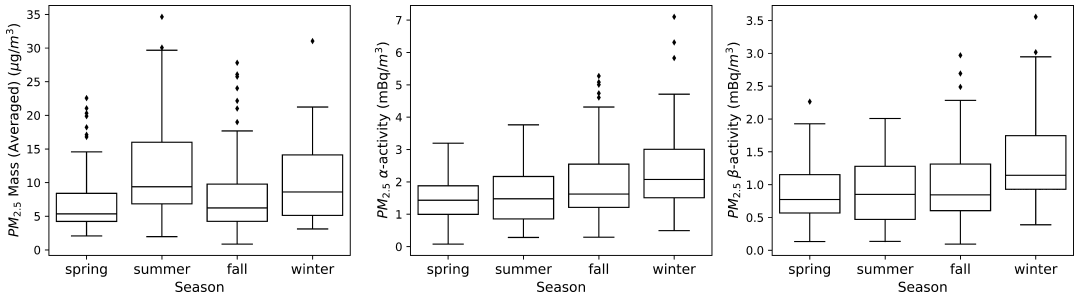

Figure 7. Boxplots of $PM_{2.5}$ mass and α- and β-activities by season.

conclusion. Physically, one would expect that high wind speed decreases particle concentrations, thus also decreasing particle radioactivity, which is the opposite of our findings. Moreover, some studies have found that Rn gas is more easily drawn from the ground during periods of decreasing atmospheric pressure.[64,65] However, based on 30 years of hourly weather model, the typical climate patterns of Boston show that the wind speed of Boston is generally higher in winter, and the dominant wind direction is from south-est to north-east.[66] This means that in winter when the wind speed is higher, the air mass arrived at Boston mainly comes from the continent, rather than the ocean. As explained before, the emanation rate of ^{222}Rn from continental regions is higher compared to the ocean. In addition, atmospheric pressure is higher during the winter season. For these reasons, it is reasonable that we found positive associations between wind speed and PR as well as between pressure and PR.

We compared the $PM_{2.5}$ α- and β-activities from the Harvard Supersite with the β-activity levels measured by the EPA RadNet at the Boston site. For security purposes, the exact location of a RadNet site within a city is not disclosed. We found a high correlation between our measurements, which are based on archived filters, and those reported by EPA RadNet, which are based on fresh filters. Furthermore, in general, the EPA RadNet β-activity measurements are lower than ours. This may be explained by three reasons. First, the analytical time of our samples and RadNet samples is not the same. As mentioned before, RadNet β-activity measurement is done 5−10 days after the air filters are collected,[42] at which point α-activities reach the minimum level due to the complete decay of short-lived ^{222}Rn progenies.[38] Following that, α-activities of the air filters gradually increase as ^{210}Pb decays to ^{210}Po that emits α-particles. Our measurement were conducted after the air filters were collected for 14 years, therefore more ^{210}Po accumulated on the air filters compared to RadNet samples. Second, as RadNet air filters were collected twice a week,[42] while our air filters were collected daily, the RadNet air filters have a higher filter loading. Few research works have looked into the relationship between the radiation counting efficiency and filter loading, but some studies investigating X-ray fluorescence on air filters stated that the accuracy of the method depends on the ideal mass loading on the filter and the ability that air mass absorbs X-rays.[67,68] Based on that, we proposed that with more particle mass building up on the filter, the particulate cake becomes thicker and less efficient in the radiation counting process, because only the α- and β-signals from the top layer particles could be fully detected. Third, our samples were collected using 37 mm Teflon membrane filters,[33] which are made of a polytetrafluoroethylene porous membrane that is flat, thin, and smooth on one side.[69] However, RadNet samples were collected on 4 inch polyester fiber filters,[42] which are thicker, rougher, and more heterogeneous compared to Teflon filters, blocking some of the radiation signals from being counted by the equipment.

Table 2. Linear Correlation Coefficient between PR and Some Meteorological Factors

variables	gross $\alpha_{n=311}$		gross $\beta_{n=296}$	
	coefficient	p-value	coefficient	p-value
$PM_{2.5}$ mass	0.06	<0.001	0.06	<0.001
temperature	−0.01	<0.01	−0.01	<0.01
pressure	0.35	<0.01	0.19	0.14
precipitation	−0.20	<0.01	−0.04	0.59
wind speed	0.09	<0.001	0.05	0.01
PBL height	-3×10^{-4}	<0.01	-5×10^{-5}	0.71
temporal proportion of back-trajectories over land	0.37	<0.001	0.43	<0.001

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■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c02284.

Linear correlation coefficient between PR and some meteorological factors; boxplots of PM_{10} mass and α- and β-activities by season; and boxplots of $PM_{2.5}$ and PM_{10} α- and β-activities by month (PDF)

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Notes

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■ ABBREVIATIONS

PM	particulate matter
PR	particulate radioactivity
LOD	limit of detection
PBL	planetary boundary layer

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Due to a production error, this paper was published ASAP on September 14, 2020, with Table 1 in the Methods section. Table 1 was moved to the Results section and the corrected paper was reposted on September 22, 2020.

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